An evaluation of ^{14}C age relationships between co-occurring foraminifera, alkenones, and total organic carbon in continental margin sediments

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Received 22 October 2004; revised 23 December 2004; accepted 25 January 2005; published 15 March 2005.

[1] Radiocarbon age relationships between co-occurring planktic foraminifera, alkenones, and total organic carbon in sediments from the continental margins of southern Chile, northwest Africa, and the South China Sea were compared with published results from the Namibian margin. Age relationships between the sediment components are site-specific and relatively constant over time. Similar to the Namibian slope, where alkenones have been reported to be 1000–4500 years older than co-occurring foraminifera, alkenones were significantly (~1000 years) older than co-occurring foraminifera in the Chilean margin sediments. In contrast, alkenones and foraminifera were of similar age (within 2σ error or better) in the NW African and South China Sea sediments. Total organic matter and alkenone ages were similar off Namibia (age difference TOC alkenones: 200–700 years), Chile (100–450 years), and NW Africa (360–770 years), suggesting minor contributions of preaged terrigenous material. In the South China Sea, total organic carbon is significantly (2000–3000 years) older owing to greater inputs of preaged terrigenous material. Age offsets between alkenones and planktic foraminifera are attributed to lateral advection of organic matter. Physical characteristics of the depositional setting, such as seafloor morphology, shelf width, and sediment composition, may control the age of co-occurring sediment components. In particular, offsets between alkenones and foraminifera appear to be greatest in deposition centers in morphologic depressions. Aging of organic matter is promoted by transport. Age offsets are correlated with organic richness, suggesting that formation of organic aggregates is a key process.

Citation: Mollenhauer, G., M. Kienast, F. Lamy, H. Meggers, R. R. Schneider, J. M. Hayes, and T. I. Eglinton (2005), An evaluation of ^{14}C age relationships between co-occurring foraminifera, alkenones, and total organic carbon in continental margin sediments, *Paleoceanography*, *20*, PA1016, doi:10.1029/2004PA001103.

1. Introduction

[2] Continental margins, on which rates of sediment accumulation are typically high, are prime locations for the study of rapid climate change [e.g., *Keigwin and Pickart*, 1999; *Shackleton et al.*, 2000; *de Garidel-Thoron et al.*, 2001; *Arz et al.*, 2003]. Many of these studies use multiple proxy parameters in order to reconstruct various aspects of past climates [e.g., *Cacho et al.*, 1999; *Herbert et al.*, 2001]. Establishment of reliable timescales is crucial. These are usually based on radiocarbon ages of planktic foraminifera [e.g., *Arz et al.*, 2003]. Foraminifera are preferred over total organic carbon (TOC) due to their unambiguous marine origin, well-known habitat, and rapid sedimentation [*Fok-Pun and Komar*, 1983]. TOC is composed of potentially variable mixtures of marine and terrestrial components, especially in sediments from continental margins. The terrestrial organic matter may have been stored in soils prior to its transport and could thus be significantly preaged upon deposition in marine sediments.

[3] With recent advances that allow dating of microgram quantities of carbon [*Pearson et al.*, 1998; *von Reden et al.*, 1998], radiocarbon analyses are now possible at the molecular level [*Eglinton et al.*, 1996, 1997]. The results can help to elucidate sources and transport pathways of organic compounds in marine sediments [*Pearson et al.*, 2000; *Pearson and Eglinton*, 2000]. Alkenones benefit from this approach. These compounds are of algal origin and can be found in most marine environments and underlying sediments [e.g., *Müller et al.*, 1998]. Notably, the abundance ratio of alkenones with different levels of unsaturation is temperature-dependent [e.g., *Brassell et al.*, 1986]. This relationship is used to reconstruct past sea surface temperatures via the $U^{K'}_{37}$ index [e.g., *Prahl et al.*, 1988], and is now widely applied in

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0883-8305/05/2004PA001103$12.00

paleoceanographic research. The alkenones are synthesized by a restricted but cosmopolitan group of marine phytoplankton. Accordingly, like planktic foraminifera, they are excellent tracers of surface water conditions.

[4] *Ohkouchi et al.* [2002] have shown, however, that radiocarbon ages of alkenones and foraminifera from identical sediment depth intervals can be offset by up to several thousand years. These authors studied sediments from the Bermuda Rise, a deep sea drift deposit in which fine-grained sediments derive predominantly from sources on the Canadian margin off Nova Scotia [e.g., *Keigwin and Jones*, 1994]. They concluded that alkenones are entrained in the advected material emanating from the Canadian margin and inferred that alkenones at the Bermuda Rise are a mixture of allochthonous and autochthonous components. Their observations demonstrate that transport of particles by currents can lead to significant spatial and temporal offsets between alkenones and co-occurring foraminifera.

[5] Sediments from the continental margin off Namibia within the Benguela coastal upwelling area have been studied in the same way [*Mollenhauer et al.*, 2003]. This area is influenced by the northward flowing eastern boundary current called the Benguela Current. Waters overlying the wide shelf are very highly productive (average chlorophyll-*a* Chl-*a*: 1.35 mg/m^3; concentrations in surface waters as determined by SeaWIFS in 1999 were used as a measure of productivity; values for individual core sites are extracted from a grid with 0.088° resolution; Table 1). Adjacent lands are arid. Inputs of terrigenous organic material are low. Alkenone ages in core top sediments from the mud lens on the inner Namibian shelf suggest rapid, in situ deposition. In contrast, alkenones extracted from a core top sample taken from the upper slope (1683 m water depth) are approximately 2500 ^{14}C years older than the co-occurring foraminifera. This age offset remains approximately constant down core. Moreover, TOC and alkenone ages agree quite well. This is interpreted as the result of continuous cycles of resuspension and redeposition of particles originating from the inner shelf.

[6] *Ransom et al.* [1998] suggest that particles bearing organic matter are repeatedly aggregated and disaggregated on the seafloor. Organic matter is thereby cycled between surface sediments and bottom water. During these cycles the aggregates could be laterally distributed and moved downslope. Foraminifera are much less susceptible to lateral transport due to their larger grain size and higher density. Bottom currents, measured on a mooring near one of the core sites, varied between 0 and 25 cm/sec and at a frequency related to internal tides. These velocities are sufficient to bring organic aggregates into suspension [*Thomsen and Gust*, 2000]. The upper continental slope off Namibia is furthermore characterized by bottom morphology with crests and valleys. The latter represent depocenters in which sediment can be focused and trapped [cf. *Mollenhauer et al.*, 2002, Figure 9]. Transport across the wide shelf, with subsequent accumulation of fine grained sediments in these depressions, is an important depositional process on the Namibian slope. The sediment near the coast consists of unconsolidated opal and organic rich deposits [*Emeis et al.*, 2004]. The aging of organic

Table 1. Core Locations and Characteristics of the Studied Sediment Cores[a]

Site	Latitude	Longitude	Water Depth, m	Annual Mean Chl-*a* Concentration, mg/m^3	Average Sedimentation Rate, cm/kyr	Average TOC Content, % dry wt.	Average Carbonate Content, % dry wt.	Climatic Setting Mode of Terrigenous Input
South China Sea, 18287-2	5.65°N	110.56°E	598	0.13	30, 60	0.7, 1.0	20, 2	humid, high fluvial input
NW Africa, GeoB5546-2	27.53°N	13.73°W	1072	0.75	20, 30	0.8, 1.6	40, 60	arid, predominantly aeolian input
Chile, GeoB3313-1	41.00°S	74.75°W	852	1.22	100	1.8	8	humid, high fluvial input
Namibia, IOW226660-5	24.11°S	12.77°E	1821	1.35	7, 15	2.2, 6.8	75, 50	arid, predominantly aeolian input

[a]Average bulk sediment data refer to Holocene (first number) and glacial (second number) core sections, respectively (T. Freudenthal, unpublished data, 2002 (NWA); *Lamy et al.* [2002] (Chile, TOC); F. Lamy et al. (unpublished data, 2000) (Chile, carbonate); *Mollenhauer et al.* [2003] (Namibia); *Steinke et al.* [2003] (SCS carbonate); *Steinke* [2001] (SCS, TOC)).

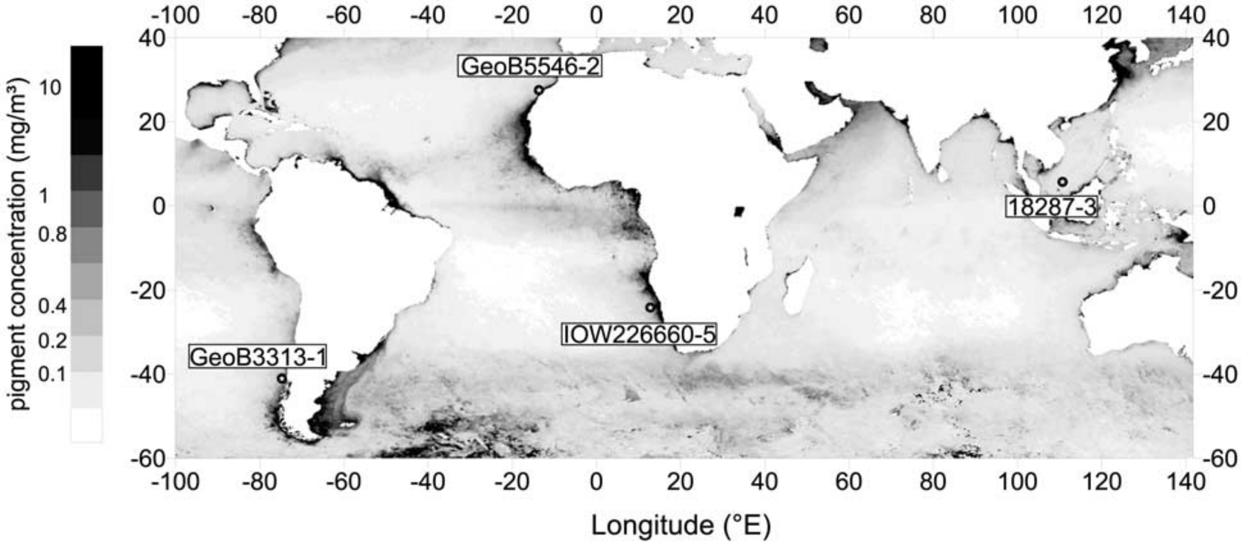

Figure 1. Locations of study sites on continental margins. Mean annual primary productivity is approximated by pigment concentrations in surface waters, given in shades of grey (pigment data from Coastal Zone Color Scanner, November 1978 to June 1986).

materials is likely to occur within the fluffy benthic boundary layers. As a result of minimal input of terrigenous material, autochthonous biogenic matter escapes rapid burial by detrital mineral grains and is therefore subject to degradation and transport processes occurring at the sediment-water interface.

[7] In sum, lateral transport can strongly control the provenance and age of sedimentary components. The resulting temporal offsets can have profound implications for interpretation of proxy records. Moreover, given the growing interest in the development of high temporal resolution paleoclimate records from rapidly accumulating sediments, development of a better understanding of the prevalence and magnitude of such offsets is imperative.

[8] Here we present new alkenone radiocarbon data for cores retrieved from high deposition rate sediments from the northwest African continental margin, the southern Chilean margin, and the South China Sea (Figure 1; Table 1). These sites differ with regard to primary productivity, climate, and seafloor morphology. Age relationships between alkenones and co-occurring foraminifera are site-specific and can be related to local depositional settings and sediment composition.

2. Study Areas

2.1. Southern Chilean Margin (SCM)

[9] The western coast of South America is largely influenced by the Peru-Chile eastern boundary current, which advects cold and nutrient-rich waters to the north. Owing to coastal upwelling north of 37°S, very high rates of primary productivity prevail along the path of this Current. The Peru-Chile Current originates from the northward flowing branch of the Antarctic Circumpolar Current (ACC), which transports subpolar waters toward the coast of South America between 40°S and 45°S, whereupon it bifurcates [*Strub et al.*, 1998].

[10] Core GeoB3313-1 (41.00°S, 74.75°W, 852 m) was taken from a small marine basin on the upper slope off southern Chile (Figures 1 and 2). This area is influenced by subpolar waters advected via the ACC and high riverine runoff causing low surface water salinities and high discharge of suspended terrigenous material [*Hebbeln et al.*, 2000]. Prevailing westerly winds suppress upwelling. The high primary productivity (Chl-a: 1.22 mg/m^3) is probably caused by mixing of iron-limited, high-nutrient, low-chlorophyll waters advected by the ACC with riverine waters supplying micronutrients such as iron [*Hebbeln et al.*, 2000]. The combination of high primary production and the discharge of large amounts of suspended load result in very high sedimentation rates, on the order of 100 cm per kyr [*Lamy et al.*, 2001, 2002]. Notably, the carbonate content of the bulk sediment is very low (Table 1), which makes it difficult to obtain sufficient foraminifera for radiocarbon dating from a small sample.

2.2. Northwest African Margin (NWA)

[11] The coast of northwest Africa is likewise influenced by an Eastern Boundary Current, the Canary Current, advecting colder and nutrient-rich waters and resulting in high primary production. Core GeoB5546-2 (27.53°N, 13.73°W, 1072 m), was retrieved off southern Morocco, to the east of the Canary Islands. The site is located north of the main upwelling system in an area which is characterized by high to moderate productivity rates (Chl-a: 0.75 mg/m^3; Figure 1). In contrast to the southern Chilean site, this core was not taken from a basin on the slope but rather from an area with parallel seismic reflectors, implying locally homogenous sedimentation without significant focusing. The shelf width at this part of the African coast is between 30 and 100 km. The adjacent coastal climate is dry. Terrigenous sediment input largely consists of Saharan desert dust [*Schütz et al.*, 1981]. However, during the wetter phase of the early Holocene, fluvial input contributed sig-

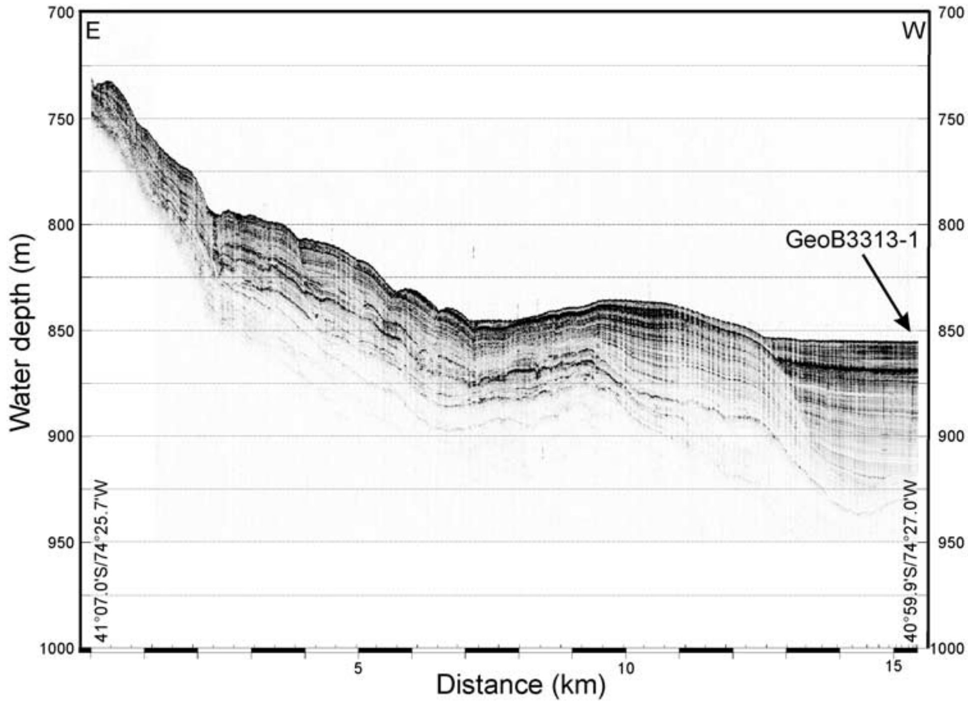

Figure 2. PARASOUND (3.5 kHz) echosounder downslope profile of the Chilean continental margin at the core site of GeoB3313 (E-W from 41°07.0s/74°25.7′W to 40°59.9′S/74°27.0′W). Parallel reflectors, deep penetration, and thickened sediment units indicate that the core was taken at a site of soft sediments and sediment focussing. Modified after *Hebbeln et al.* [1995].

nificantly to terrigenous sediment flux (H. Kuhlmann et al., Meridional shifts of the Mediterranean and monsoonal climate system off NW Africa during the last 130,000 years: Implications from river discharge, submitted to *Quaternary Research*, 2005). Prevailing surface currents flow southward along the shore line. Particles in surface waters are also transported by eddies and upwelling filaments [*Freudenthal et al.*, 2001, and references therein]. Less intense, deep water currents are also directed southward.

2.3. South China Sea (SCS)

[12] The South China Sea (SCS) is a large marginal basin in the western Pacific influenced by the Western Pacific Warm Pool and the Asian monsoon system. Its location between the east Asian land mass and the western Pacific makes the SCS very sensitive to both terrestrial and oceanic climate change. Modern hydrography in the SCS is controlled by seasonal inflow of western Pacific water from the northeast. In contrast to the surface waters overlying other core sites on continental margins, the SCS is oligotrophic (Chl-a: 0.13 mg/m^3) and sedimentation is dominated by terrigenous detritus supplied through some of the world's largest rivers [*Milliman and Meade*, 1983]. The high rates of siliciclastic sedimentation are significantly enhanced during glacial periods when large shelf areas were exposed during sea level lowstands [*Pelejero et al.*, 1999]. Concordant development of additional drainage systems further increased terrigenous input, particularly in the southwestern SCS [*Schönfeld and Kudrass*, 1993; *Wang et al.*, 1999].

[13] Core 18287-3 (5.65°N, 110.65°E, 598 m) was retrieved from the upper continental margin of the southern part of the SCS adjacent to a very large shelf area, the Sunda Shelf. This core location is near the mouth of the glacial Molengraaff River, which drained the emergent shelf during sea level lowstands. High glacial sedimentation rates were sustained by high fluxes of both siliciclastic and organic terrigenous detritus. Sedimentation rates declined in response to deglacial flooding of the shelf (see *Steinke et al.* [2003] for more details on age model and sedimentology).

3. Methods

[14] The existing data set [*Mollenhauer et al.*, 2003] from off Namibia consists of corresponding radiocarbon ages of planktic foraminifera, bulk organic carbon (TOC), and alkenones. In order to draw comparisons with new data, samples from the SCM, SCS and NWA sites were chosen at or near depths for which radiocarbon ages of planktic foraminifera had already been obtained [*Kienast et al.*, 2001; *Lamy et al.*, 2001; *Kuhlmann et al.*, 2004]. Radiocarbon ages of TOC and alkenones in these samples were then determined. Standard methods were used for preparation of TOC samples [e.g., *McNichol et al.*, 1994]. Alkenone purification requires dedicated techniques for wet chemical separation [*Xu et al.*, 2001]. A detailed description of the method is given in the work of N. Ohkouchi et al., Radiocarbon dating of alkenones from marine sediments: I. Isolation protocol, submitted to *Radiocarbon*, 2004, here-

inafter referred to as Ohkouchi et al., submitted manuscript, 2004). In brief, 50–150 g of freeze-dried sediment were solvent extracted for 48 hours in a large-volume Soxhlet apparatus using dichloromethane and methanol (93:7, ~750 mL). The extracts were reduced to dryness and saponified using 0.5M KOH in methanol (~15 mL, 80°C, 2 hours). The products were extracted with *n*-hexane after addition of distilled water. This extract was separated into three fractions using small columns filled with silica gel (100–200 mesh, 5% deactivated). Ketones were eluted in the second fraction with dicholoromethane:hexane (2:1) and further purified by urea adduction. The urea adduct was then passed through a column filled with $AgNO_3$-impregnated SiO_2 to isolate unsaturated ketones. A final step of silica gel chromatography removed any remaining impurities from the alkenone fraction. The resulting fraction contains C_{36}-C_{39} alkenones in high purity (Ohkouchi et al., submitted manuscript, 2004). The samples were transferred in solvent to precombusted quartz-glass tubes. The solvent was eliminated under a stream of nitrogen, precombusted copper oxide was added, and the tubes were evacuated and flame-sealed. Samples were combusted overnight (850°C), and the resulting CO_2 was stripped of water and quantified. The carbon dioxide was subsequently converted to graphite by reduction over a cobalt catalyst in the presence of hydrogen for radiocarbon analysis by accelerator mass spectrometry. Analyses were performed at the National Ocean Science AMS (NOSAMS) facility at the Woods Hole Oceanographic Institution. AMS measurements of small samples containing less than the generally required amount of 0.5–1 mg of carbon require dedicated techniques [*Pearson et al.*, 1998; *von Reden et al.*, 1998].

[15] Entrainment of minor amounts of carbon from unknown and potentially variable sources is an inescapable consequence of the preparation and combustion procedure. This "carbon blank" [*Pearson et al.*, 1998] is of increased importance in processing small samples, and its contribution to the resulting CO_2 must be carefully assessed. This is reflected in larger error margins as reported for small samples than for regular sized samples such as foraminifera or TOC. An evaluation of potential blanks associated with the alkenone purification method is discussed separately (G. Mollenhauer et al., Radiocarbon dating of alkenones from marine sediments: II. Assessment of carbon process blanks, submitted to Radiocarbon, 2004, hereinafter referred to as Mollenhauer et al., submitted manuscript, 2004). Blanks were found to be negligible compared with the combustion blank as found by NOSAMS in a separate study [*Gagnon et al.*, 2003]. Uncertainties in the ages of small sample of alkenones increase with sample age, but accuracy is generally better than 500 years in samples younger than approximately 10,000 years (G. Mollenhauer et al., submitted manuscript, 2004).

[16] Radiocarbon levels are reported as conventional radiocarbon ages. Accordingly, they have been corrected for isotopic fractionations that have occurred during the formation of the dated material, or which occurred during processing and measurement [see *Stuiver and Polach*, 1977]. Thus, despite the different $\delta^{13}C$ values of organic matter and carbonates, this convention allows for direct comparison of radiocarbon contents of foraminifera, alkenones and TOC.

4. Results

[17] Results of radiocarbon measurements are summarized in Figure 3 and Table 2. In Figure 3, data for one of the two Namibian cores (IOW226660-5, 24.11°S, 12.77°E, 1821 m) from *Mollenhauer et al.* [2003] are included for comparison. Radiocarbon ages increase with depth in all cases. Moreover, the slopes of the respective age-depth relationships are characteristic for each core and similar for all dated fractions. In these cores, sedimentation rates could be derived from radiocarbon analyses of any of these fractions. In the South China Sea core, TOC ages are on average 2200 ^{14}C years older than those of foraminifera from the same depth (or the interpolated foraminiferal age at the same depth). Ages of alkenones, in contrast, agree well with those of foraminifera considering the added uncertainty associated with combustion-blank corrections for particularly small samples (the maximum difference is 700 years). In the NW African core, ages of all sediment fractions agree within a few hundred years. For the alkenones measured as "small samples" (i.e., <300 μg carbon) this is within the error of age determination. The TOC samples are slightly older than the corresponding foraminifera; both TOC and foraminifera were dated as large samples (containing at least 300 μg C). The Chilean core data are very similar to those obtained off Namibia [*Mollenhauer et al.*, 2003]. Age offsets between the foraminifera and both TOC and alkenones, which themselves agree quite well, are significant and constant. The only significant difference between the Namibian and the Chilean sites is the magnitude of the age offset, which is on the order of 1000 ^{14}C years off Chile, while they average 2500 ^{14}C years off Namibia.

5. Discussion

[18] There are several potential causes for the observed age offsets. The most plausible are differential bioturbation of particles of different size [e.g., *Bard*, 2001] and advection of resuspended material [cf. *McCave and Tucholke*, 1986;

Figure 3. Radiocarbon dating results for planktic foraminifera (circles), alkenones (diamonds), and bulk OC (squares) for the three cores studied here. For reference, results for one of the cores studied in *Mollenhauer et al.* [2003] are also given (IOW226660, Namibia). Upper panels show conventional radiocarbon ages versus core depth. Error bars denote 1σ errors. Radiocarbon age differences between alkenones and foraminifera (downward pointing triangles) and between bulk OC, and foraminifera (upward pointing triangles) are plotted on the lower panels. For the South China Sea core 18287-3, age differences are calculated as difference to interpolated foraminifera age at depths where no corresponding foraminiferal age was available (indicated with asterisks). Radiocarbon ages of foraminifera are from *Lamy et al.* [2001] (Chile), *Kienast et al.* [2001] (SCS), and *Kuhlmann et al.* [2004] (NW Africa).

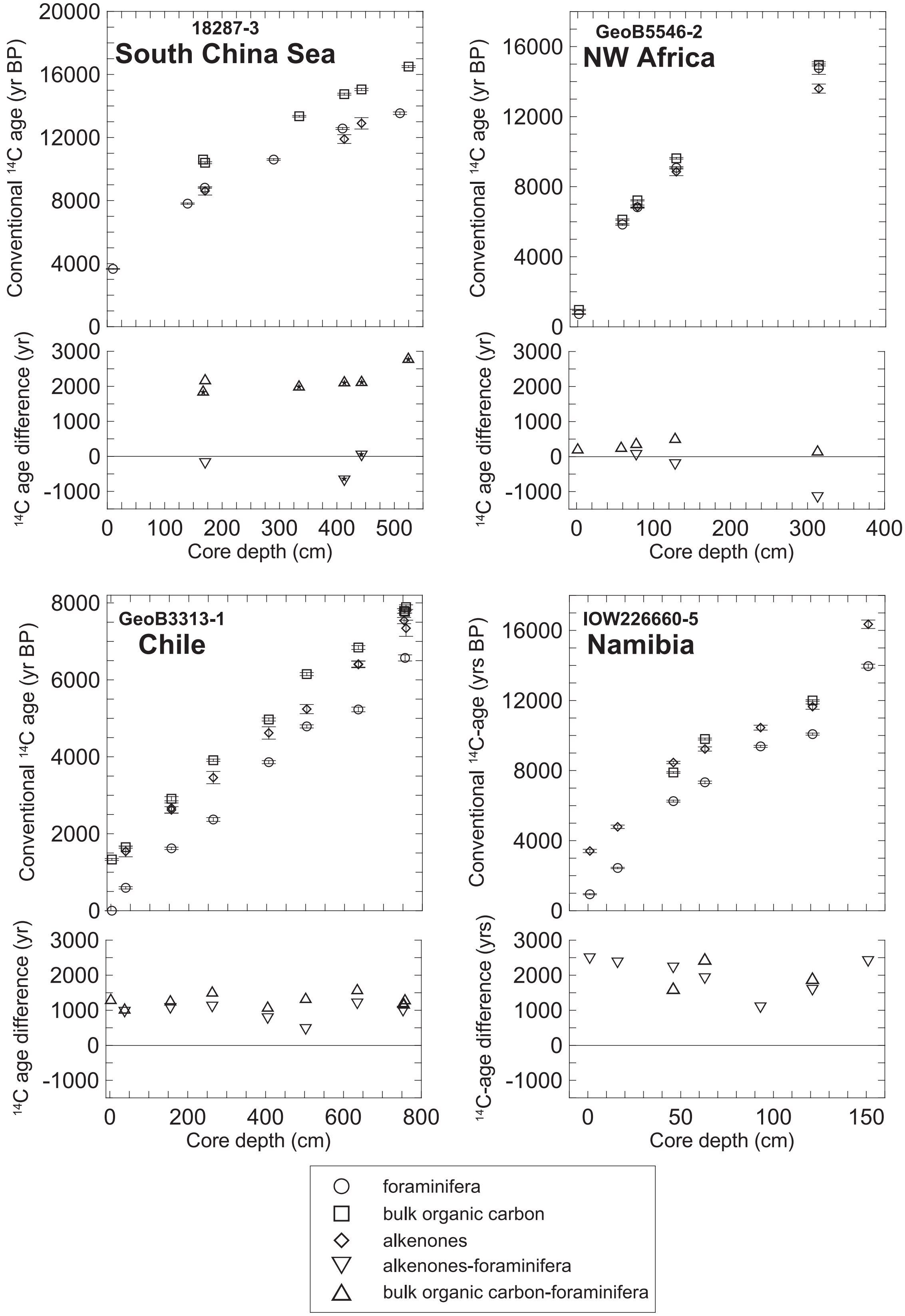
18287-3
South China Sea
GeoB5546-2
NW Africa
GeoB3313-1
Chile
IOW226660-5
Namibia
Conventional ¹⁴C age (yr BP)
¹⁴C age difference (yr)
Conventional ¹⁴C-age (yrs BP)
¹⁴C-age difference (yrs)
Core depth (cm)
foraminifera
bulk organic carbon
alkenones
alkenones-foraminifera
bulk organic carbon-foraminifera

Figure 3

Table 2. Radiocarbon Contents of TOC and Alkenone Samples[a]

Sample	Foraminifera		Bulk OC		Alkenones		
	fMC	Age, ^{14}C years BP	fMC	Age, ^{14}C years BP	fMC	Age, ^{14}C years BP	Sample Size, μg C
South China Sea, 18287-2							
165–169 cm (167 cm)		*(8715)*	0.2665 ± 0.0016	10600 ± 45			
169–172 cm (170 cm)		*8815 ± 40*	0.2733 ± 0.002	10400 ± 60	0.3424 ± 0.0113	8610 ± 260	48
332–336 cm (334 cm)		*(11310)*	0.1892 ± 0.0012	13350 ± 50			
411–415 cm (413 cm)		*(12600)*	0.1598 ± 0.0012	14750 ± 60	0.2271 ± 0.0081	11900 ± 280	32
441–445 cm (443 cm)		*(12890)*	0.1536 ± 0.0013	15050 ± 70	0.2009 ± 0.0092	12900 ± 360	26
523–527 cm (524.5 cm)		*(13690)*	0.1283 ± 0.001	16500 ± 60			
NW Africa, GeoB5546-2							
0–3 cm	0.9139 ± 0.0032	725 ± 25	0.8865 ± 0.0041	965 ± 35			
56–61 cm (58 cm)		*5835 ± 40*	0.4669 ± 0.0023	6120 ± 40			
75–80 cm (78 cm)		*6825 ± 15*	0.4072 ± 0.0018	7220 ± 35	0.4257 ± 0.0049	6860 ± 90	135
126–131.5 cm (128 cm)		*9080 ± 70*	0.3020 ± 0.0018	9620 ± 45	0.3325 ± 0.0093	8850 ± 220	200
311–316 cm (313 cm)	0.1591 ± 0.0002	14770 ± 360	0.1558 ± 0.0011	14950 ± 55	0.1844 ± 0.0060	13600 ± 260	120
Chile, GeoB3313-1							
1–4 cm	1.0610 ± 0.0044	modern	0.8471 ± 0.0029	1330 ± 25			
36–40 cm (38 cm)		*595 ± 30*	0.8145 ± 0.0028	1650 ± 25	0.8258 ± 0.0143	1540 ± 140	66
153–158 cm		*1620 ± 30*	0.6962 ± 0.0047	2910 ± 55	0.7176 ± 0.0116	2670 ± 130	261
					0.7215 ± 0.0075	2620 ± 85	254
261–265 cm (258–268 cm)		*2370 ± 50*	0.6143 ± 0.0024	3910 ± 30	0.6504 ± 0.0133	3460 ± 160	189
404–407 cm (403–408 cm)		*3860 ± 40*	0.5384 ± 0.0026	4970 ± 40	0.5629 ± 0.0114	4620 ± 160	196
505–510 cm (503–513 cm)		*4790 ± 40*	0.4649 ± 0.0022	6150 ± 40	0.5207 ± 0.0076	5240 ± 120	97
633–638 cm		*5230 ± 60*	0.4267 ± 0.0027	6840 ± 50	0.4503 ± 0.0044	6410 ± 80	216
					0.4509 ± 0.0048	6400 ± 85	209
752–754 cm			0.3789 ± 0.0028	7790 ± 60	0.3914 ± 0.0040	7540 ± 80	168
754–757 cm (753–758 cm)		*6570 ± 80*	0.3801 ± 0.0016	7770 ± 35			
757–759 cm			0.3745 ± 0.0028	7890 ± 60	0.4011 ± 0.0108	7340 ± 210	126

[a]Given as fMC = fraction modern carbon [*Stuiver and Polach*, 1977]. Foraminiferal ages (in italics) are from *Kienast et al.* [2001] (South China Sea), *Kuhlmann et al.* [2004] (NW Africa), and *Lamy et al.* [2001] (Chile). Foram ages in parentheses are interpolated values. Depths in parentheses refer to foraminifera samples, if different from organic matter samples.

Benthien and Müller, 2000]. The first is expected to carry fine particles to greater depths in the sediment, causing organic matter to be younger than co-occurring foraminifera [*Bard*, 2001]. In contrast, redeposition of advected fine particles would cause organic sediment constituents to be older than foraminifera (see *Mollenhauer et al.* [2003] for details). Different species of planktic foraminifera were analyzed in each core. In each case, the most abundant species were chosen and shallow living species were preferred. This selectivity, together with the high rates of sedimentation at all sites, should further minimize the potential for variable ages resulting from abundance variations and bioturbation [e.g., *Broecker et al.*, 1999].

[19] Advection of fine particles, in particular on continental margins, has been suggested as an important process leading to age discrepancies in previous studies. For example, *Benthien and Müller* [2000] reported anomalously low alkenone-based SST values from core top sediments in the Argentine Basin. This discrepancy was interpreted as resulting from advection of alkenone-bearing particles from the Southern Ocean. *Freudenthal et al.* [2001] inferred advection of fine-grained particles from the nitrogen isotopic composition and relative trapping efficiencies in shallow and deep sediment traps off NW Africa. Similar results were reported from a sediment trap study of alkenone fluxes in the Norwegian Sea [*Thomsen et al.*, 1998]. Finally, *Lampitt et al.* [1995] concluded from oxygen consumption data that downslope transport of sediment on the northeast Atlantic continental margin plays a significant role in sedimentation processes. Selective advection of fine-grained particles can mix organic matter and foraminifera with differing ages and sources. Compound-specific radiocarbon dating of marine biomarkers provides a tool to study potential temporal decoupling, as two types of marine material deriving from surface water are compared.

5.1. South China Sea

[20] The South China Sea provides an example in which the placement of multiple marine proxies on a single age scale appears robust. Radiocarbon ages of planktic foraminifera and alkenones in core 18287-2 agree quite well. This supports the inferences made by *Kienast et al.* [2001], who found in the same core that an age scale derived from AMS dates of planktic foraminifera led to an alkenone-based SST record that correlated well with Greenland ice core data. We can now demonstrate that, for this particular core, the assertion of coeval deposition of foraminifera and alkenones is valid. Accordingly, the observed correlation is a real feature that can be interpreted in a paleoclimatic context. A more detailed discussion of the alkenone ages from the SCS can be found in the work of M. Kienast et al. (manuscript in preparation, 2005).

[21] In contrast to the alkenones, TOC and foraminiferal ^{14}C ages in core 18287 are offset by 2000 to 3000 radiocarbon years. This offset between the marine markers (i.e., foraminifera and alkenones) and TOC is plausibly due to inclusion of preaged terrigenous organic matter in the total organic carbon. Studies in tropical estuaries have shown that

much of the terrigenous organic matter is degraded very rapidly upon deposition on marine shelves [*Aller*, 1998; *Aller and Blair*, 2004]. Only the very refractory material, which has the lowest radiocarbon content, remains. If the terrigenous material was radiocarbon dead at the time of deposition, and the Δ^{14}C of the marine organic end-member was equal to that of the foraminifera, preaged terrigenous material accounted for 21 to 30% of the sedimentary TOC. The larger relative contributions pertain to the older sediments. This estimate of terrigenous inputs is supported by the $\delta^{13}C_{org}$ record from the same core, which indicates a terrigenous (C_3 vascular plant) origin of a fraction of the bulk organic matter at this site [*Steinke et al.*, 2003], and with n-alkane records from throughout the SCS [*Pelejero*, 2003], all of which indicate that delivery of terrigenous organic matter was higher in glacial intervals.

[22] The South China Sea is an area in which local primary productivity is relatively low. Consequently, total organic carbon contents in the sediment are low. The high input of terrigenous material results in the relatively high sedimentation rates (20–60 cm/kyr), which may be responsible for a rapid burial of all marine organic matter. If marine organic matter is reaching the seafloor largely as fecal pellets or other rapidly sinking particles, the chances of resuspension and successive transport by currents would be reduced.

5.2. NW Africa

[23] In core GeoB5546-2, ages of all dated sediment fractions agree remarkably well. The sediments are characterized by low concentrations of TOC and moderate contents of carbonate (0.8–1.6% dry wt. and approximately 40–60% dry wt., respectively; T. Freudenthal, University of Bremen, unpublished data). In spite of the moderate local productivity and low organic carbon content, high sedimentation rates (20–30 cm/kyr) are maintained in both Holocene and glacial intervals. In this area, terrigenous input is dominated by Saharan dust [e.g., *Schütz et al.*, 1981]. Dust collected in the core vicinity contains 0.7 to 0.9% dry weight of organic carbon [*Lavik*, 2001; *Eglinton et al.*, 2002]. TOC is slightly older than corresponding foraminifera, an offset explainable by minor contributions of preaged terrestrial organic matter. Alternatively, the terrigenous organic matter could be only slightly preaged at the time of deposition. Individual organic components of dust collected off NWA have ages ranging from 650 to 2070 radiocarbon years [*Eglinton et al.*, 2002]. The proportion of terrigenous organic matter in bulk OC would likely be enhanced by preferential degradation of marine OC. This effect has been noted for organic matter in the Madeira Abyssal Plain [*Prahl et al.*, 2003]. Stable carbon isotopic values of TOC ($\delta^{13}C_{TOC}$) averaging −19.5‰ as reported by NOSAMS (not shown), however, point to small relative contributions of terrigenous organic matter at the NWA margin, assuming dominance of C3 plants.

[24] Molluscan- and algal-carbonate sands are common on the shallow NWA shelf [*Fütterer*, 1983]. The carbonate content decreases rapidly downslope, and reaches minimum values at depths of 1000–3000 m. This pattern has been attributed to physical factors (oceanic currents and waves) rather than to productivity [*Fütterer*, 1983].

[25] The equality of ages for all dated sediment fractions suggests rapid burial of fine-grained material on the slope. Even though sediment transport seems to play a role, as suggested in previous studies [*Fütterer*, 1983; *Freudenthal et al.*, 2001], it results in no significant age offsets. Sediment focusing thus must be essentially syndepositional. As a result, the sources of sediment are probably local or regional, which implies that paleoceanographic signals can be interpreted as regional, coeval records.

[26] Alkenones from 311–316 cm sediment depth are one exception to the above trends. They are 1170 ^{14}C years younger than the co-occurring foraminifera and 1300 ^{14}C years younger than the TOC from the same sample. This may be due to contamination of this particular sample with modern carbon or with carbon younger than the dated age of 13600 ^{14}C years. Less than 10% of a "half modern" contamination and less than 3% of a "modern" contamination would be sufficient to induce such an offset. Bioturbation cannot be excluded as the cause of the alkenones being younger than the foraminifera, but it is unlikely because bioturbation should affect all organic material, including TOC, to a similar extent. The contribution of a carbon blank is always possible and, as mentioned above, more strongly affects samples that contain less carbon, i.e., old or very small samples. The alkenone sample from 311–316 cm core depth contained approximately 120 μg carbon and it was taken from a depth in the core corresponding to early deglacial times. Since it is the smallest and the oldest of the three dated alkenone samples from core GeoB5546-2 it is most sensitive to such effects. Although contamination of small samples is always a possibility, and although such contributions become more significant with sample age, it does not mean that biomarkers older than ~16,000 ^{14}C years cannot be dated reliably. In fact, very consistent dates have been generated for the Namibian cores down to a foraminiferal age of 22630 ^{14}C years [*Mollenhauer et al.*, 2003].

5.3. Chile

[27] The last of the sites considered here, GeoB3313-1, is similar to the Namibian sites. The core was taken from a high-productivity area, and was retrieved from a basin on the continental slope in which sediment accumulation rates are very high. The morphology and the sediment echogram are suggestive of sediment focusing in this basin (Figure 2). The major difference to the Namibian margin is that the core was taken from a very humid climate zone. Large amounts of terrigenous material are delivered from the adjacent continent. As a result, sedimentation rates approach 100 cm/kyr. From the depositional setting and the observations to date [*Mollenhauer et al.*, 2003], a significant age offset between organic matter and foraminifera might be expected.

[28] Indeed, both alkenones and bulk organic matter are systematically 1000 to 1500 ^{14}C years older than the co-occurring foraminifera. Constant admixture of radiocarbon dead material could cause the offset between foraminifera and organic matter ages in this core. However, no potential source for such an admixture, which would have to be rich in organic matter, can be identified. Continental rocks in this area are largely metamorphic, the shelves are sites of sedimentation rather than erosion, and sediment

hiatuses on the continental slope are only known for the Holocene (Dierk Hebbeln, University of Bremen, personal communication). Therefore constant admixture of fossil material is an unlikely explanation for the age offsets.

[29] Unlike in the South China Sea, where bulk organic matter appears to be more strongly influenced by terrigenous input, there is no clear and significant difference in age between alkenones and bulk organic matter. Two explanations are possible: First, the recalcitrant terrigenous organic fraction, while still present, contributes insignificantly to the TOC, which is on the order of 2% dry weight in this high productivity area [*Lamy et al.*, 2002]. Alternatively, terrigenous organic matter discharged into the ocean could be relatively young, indicating rapid transport with short residence times on land. This is in agreement with a rough topography and sparse vegetation [cf. *Blair et al.*, 2004], in contrast to the tropical setting adjacent to the SCS. Radiocarbon analyses on vascular-plant biomarkers would be required to confirm this interpretation.

[30] Low variability in the grain-size distribution and in the content of fine-grained siliciclastic fraction of the sediment (silt and clay) indicates that the sedimentary setting is influenced by hemipelagic processes with little variability over time [*Lamy et al.*, 2001], consistent with our observation of relatively constant age offsets. In this core, however, only Holocene sediments were analyzed. It remains possible that, during glacial times, the depositional setting may have been markedly different, resulting in different age relationships. The relatively young alkenone age at a core depth of 505–510 cm may again be caused by contamination. This sample contained 97 µg of carbon, and most of the C_{37} alkenones were lost during the purification process. This particular measurement must therefore be interpreted cautiously.

[31] In spite of the low variability in relative abundance of the fine-grained siliciclastic constituents in core GeoB3313-1, distinct paleoclimatic changes have been inferred to have taken place over the course of the Holocene [*Lamy et al.*, 2001, 2002]. Pronounced variability in regional climate has been linked to the position of the southern westerlies and suggested to also be mechanistically linked to climate variability in other regions of the World. The reported age offsets do not seem to vary significantly over the timescales in question. In contrast, they are remarkably constant over time.

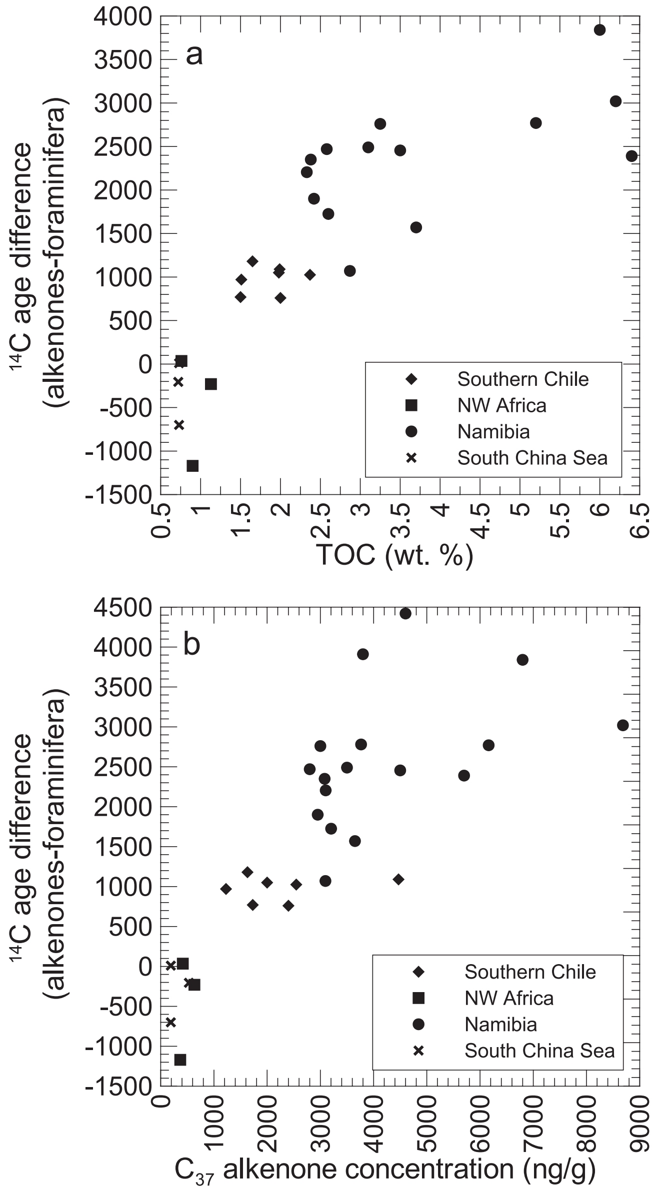

Figure 4. Correlation of (a) TOC contents and (b) C_{37} alkenone concentrations with radiocarbon age difference between alkenones and foraminifera (given in radiocarbon years) in sediments from four different continental margin settings. Namibian data are from two individual sediment cores [*Mollenhauer et al.*, 2003]. Negative age differences indicate alkenones younger than foraminifera, which may be caused by contamination of the alkenone sample.

5.4. Comparison of All Sites

[32] Constant age offsets have also been observed off Namibia [*Mollenhauer et al.*, 2003]. In those cores, similar age offsets persisted through Holocene, deglacial and even glacial sediments. In both settings, sedimentation seems to be predominantly determined by the seafloor morphology. Our data suggest that the processes leading to focusing of preaged organic matter in local basins on the continental slope are not significantly influenced by sea level or climate. Deep sea currents acting as compensation for wind-driven surface currents would be expected to respond to climatic changes. In contrast, internal tides result from gravitational forces of the sun and moon in interaction with basin geometry rather than from climatic processes. Our data may therefore indicate that resuspension processes on continental margins are independent of climatic change and a function of local sedimentological control. Strong coupling between sediment distribution and internal tides has been suggested [*Cacchione et al.*, 2002]. By extension, our radiocarbon data for the two sites off west Africa suggest a strong physical control on age relationships between sedimentary components as well. Unlike the Namibian coast, where age offsets between alkenones and foraminifera are

on the order of 2500 years, there is no shallow water mud lens on the shelf off NWA [*Fütterer*, 1983], an area where all sediment constituents show very similar ages. Moreover, the Namibian shelf is relatively deep and wide compared to the NWA shelf. This implies that shelf depth and width may in part control deposition patterns and postdepositional redistribution on the adjacent slopes.

[33] Namibian slope sediments contain 2–6.5% dry weight TOC and 2.8–8.7 μg/g C_{37} alkenones [cf. *Mollenhauer et al.*, 2003], sediments on the Chilean slope average 1.5–2.5% dry weight TOC and 1.2–4.5 μg/g C_{37} alkenones [*Lamy et al.*, 2002], while both NWA and SCS sediments contain 0.5–1.5% dry weight TOC (Freudenthal, unpublished data, Steinke, unpublished data) (Table 1). NWA and SCS sediments contain 0.3–0.6 μg/g and 0.2–0.5 μg/g alkenones, respectively. Age offsets between alkenones and foraminifera are largest where TOC content and alkenone concentration are highest, and seem to be negligible where sediments are relatively poor in TOC and alkenones (Figure 4). A possible cause for this trend would be that organic aggregates with large surface areas that are easily brought in suspension are most prevalent in high TOC settings, promoting repeated periods of resuspension. In contrast, organic matter in low TOC environments may be more strongly bound to mineral surfaces and would be buried faster along with the host mineral grains. Further studies are required to explore the precise modes of association and transport of organic matter.

[34] A detailed study of the remineralization of organic compounds in sediments off Papua New Guinea [*Aller and Blair*, 2004] has shown that preferential reaction of relatively young fractions of the TOC pool leads to ^{14}C depletion (i.e., older apparent ages) of the residual sedimentary TOC in frequently reworked shelf deposits. This type of preferential decomposition may be most effective in organic-rich deposits where macrobenthic and microbial communities responsible for efficient remineralization are abundant. In contrast, remineralization may not be as intense in sediments lean in TOC, and sites with low contents of TOC cannot support rich and abundant benthic communities. As a result, larger proportions of young marine organic matter may be preserved in low TOC sediments. Systematic studies of relative preservation potentials for various marine and terrigenous organic compounds are necessary in order to understand these processes. Clearly, more research is necessary to resolve the observed connection between age offsets and bulk OC content.

6. Conclusions

[35] Age relationships between co-occurring foraminifera, alkenones, and TOC in sediments from four different continental margin settings are site-specific and relatively constant over time. The magnitudes of the offsets depend on factors such as productivity, seafloor morphology, terrigenous input, and sedimentation rate. The mechanisms controlling age offset do not vary between climatic stages and are thus primarily determined by physical interactions between deep water currents, sediment grains, and seafloor morphology. Our observations suggest that high deposition rate environments on continental margins associated with high organic carbon contents occur at sites where currents focus fine-grained sediment. The strong correlation between age offsets and organic carbon content implies that sediment composition is an important factor as well. Continuous cycles of re-suspension and re-deposition of organic-rich aggregates, with ultimate deposition in depressions on the continental slope, may be responsible for large (up to 4500 years) age discrepancies between foraminifera and alkenones. Even in a very high sedimentation rate setting such as off Chile (100 cm/kyr), offsets of up to ~1200 years may result from continuous re-suspension/re-deposition cycles. In contrast, sediments lean in organic matter are apparently less susceptible to re-suspension of organic-rich aggregates, which in turn promotes more rapid burial of alkenones. Sediment focusing in morphologic depressions appears to be conditional for the temporal decoupling of foraminifera and alkenone records. Transport across a broad shelf may further increase the potential of significant age offsets. In contrast, rapid burial by siliciclasitc grains may prevent significant preaging of marine organic matter. The underlying processes, however, are not fully understood yet.

[36] We need to develop the capability to predict possible age offsets between sedimentary components at a specific site based on the sedimentary setting. This may prove to be a prerequisite before embarking on detailed reconstructions of paleoclimate based on information from multiple proxies, including foraminifera and biomarkers. An understanding of the underlying processes when such offsets are observed will provide valuable information regarding sources and pathways of organic matter in marine sediments. Meanwhile, phase relationships between proxy records based on different size fractions should be interpreted with caution, unless good age control exists for each size fraction. Proxy records from sediment drifts and drift-like settings on continental margins are particularly likely to be affected by transport processes.

[37] **Acknowledgments.** We thank Li Xu, Daniel Montluçon and NOSAMS staff for sample preparation and laboratory assistance. Alex Schimanski and Karl Stattegger generously provided sample material from the South China Sea. The captains and crews of R/V *Sonne* and R/V *Meteor* are acknowledged for assistance during sample collection. Eva Calvo and Liz Sikes are thanked for constructive reviews, which helped improving the manuscript. GM and MK acknowledge financial support from the WHOI postdoctoral scholarship program. This work was funded by NSF grant OCE-0327405. This is WHOI publication 11302.

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